COLLINEAR LASER PHOTOIONIZATION OF HELIUM ISOTOPES IN A FAST ATOMIC BEAM

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We studied laser isotope-selective two-step photoionization of metastable helium atoms in a fast beam. The detection selectivity attained for the rare isotope ^{3}He was 10^6.

1. Introduction

The development of laser methods of detecting single atoms [1] has made it possible to set out to perform an ultrasensitive isotope analysis of substances containing rare isotopes in concentrations of less than 10^{-10} of that of the main isotope [2]. Several methods based on the isotope-selective action of laser radiation have been proposed for detecting such isotopes. These include the multistep photoionization of atoms [3], fluorescence of atoms in a beam [4] and at the exit from an ordinary mass spectrometer [5], stepwise collinear photoionization of atoms in a fast beam [6], resonant laser depletion of a preliminarily excited state [7], magnetic mass selection in conjunction with resonant neutralization and laser resonant ionization [8,9], and selective deceleration [10] and deflection [11] of atoms.

Considerable progress has recently been made in detecting rare radioactive Kr isotopes with the use of preliminary enrichment and resonant ionization spectroscopy techniques [12], in studying the optical characteristics of short-lived isotopes by means of collinear laser spectroscopy [13] and resonant ionization mass spectrometry [14,15], and also in detecting isotopes with the aid of resonant excitation and ionization by collision in thermionic diode detectors [16].

In this paper we present the results of experimental studies of collinear laser photoionization of fast helium atoms. The idea of the method is that the isotope-selective stepwise excitation and ionization of the atoms being detected are effected in an accelerated atomic beam obtained by neutralizing an ion beam preliminarily accelerated in a given accelerating potential U_A [6]. Along with the high sensitivity inherent in photoionization detection techniques [17], this method possesses the following important advantages.

(i) The method provides for an additional isotope shift in atomic absorption spectra on any atomic transition.

(ii) It ensures the narrowing of the Doppler-broadened spectral lines down to the radiative linewidth and thus allows narrow-band laser radiation to interact with all the atoms of the accelerated beam [18].

(iii) The method makes it possible to carry out, using the existing dye lasers, isotope-selective excitation to Rydberg states of atoms having a high ($\simeq$20 eV) ionization potential and featuring high-lying metastable states.

The experiments on the two-step excitation of potassium isotopes to Rydberg states demonstrated the possibility of achieving a detection selectivity of ^{40}K as high as 10^5 [19]. In that case, the isotope-selective excitation was effected in a single step only.

The next step toward radical improvement of detection selectivity would naturally be the use of the effect of multiplication of excitation selectivities achieved at each step, e.g., $S=S_1\times S_2$ for two-step excitation [3]. The maximum selectivity of excita-

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tion of a rare isotope by means of a laser radiation with a bandwidth of $\Delta\nu_L$ satisfying the condition $\Gamma < \Delta\nu_L \ll \Delta\nu_i$, where Γ is the homogeneous absorption line halfwidth and $\Delta\nu_i$ the isotope shift, is given by the expression $S_{1,2} = (\Delta\nu_i/\Gamma)^2(2\Gamma/\Delta\nu_L)$ [#1]. For example, in the case of helium, the homogeneous halfwidth of absorption lines on the transitions considered later in the text is governed by the lifetime of the $3\,^3P$ level and is $\Gamma = 0.84$ MHz. The selectivities of the excitation of the ^{3}He isotope at the first and second excitation stages at $\Delta\nu_L = 0.5$ cm^{-1} are 7.2×10^6 and 1.4×10^6, respectively. The total spectral selectivity S of the above two-step excitation process is then 10^{13}.

There are, however, a number of collisional processes which must be considered when estimating the ultimate selectivity of detection of a rare isotope. To study these processes, we selected the isotope ^{3}He the natural abundance of which is 1.4×10^{-6} of that of the main isotope ^{4}He.

The ionization potential of the helium atom (fig. 1) is $E_i = 24.6$ eV and therefore, to effect its stepwise ionization starting from the ground state and using the existing lasers is impossible. The helium atom has two long-lived metastable states: the $2\,^1S_0$ singlet state and $2\,^3S_1$ triplet state. Using the phenomenon of charge exchange between accelerated ions and alkali metal vapors, it is possible to prepare fast neutral atoms in these metastable states. Potassium atoms are a suitable candidate for such an exchange. The electron-binding energy in the $2\,^1S$ and $2\,^3S$ states of the He atom is close to the ionization energy of K from its ground state $4S_{1/2}$. As a result of charge exchange, approximately 3/4 of the atoms are formed in the triplet metastable state $2\,^3S$ [21]. The isotope-

[#1] More exact expressions for many cases are given in refs. [17] (Chap. 9) and [20].

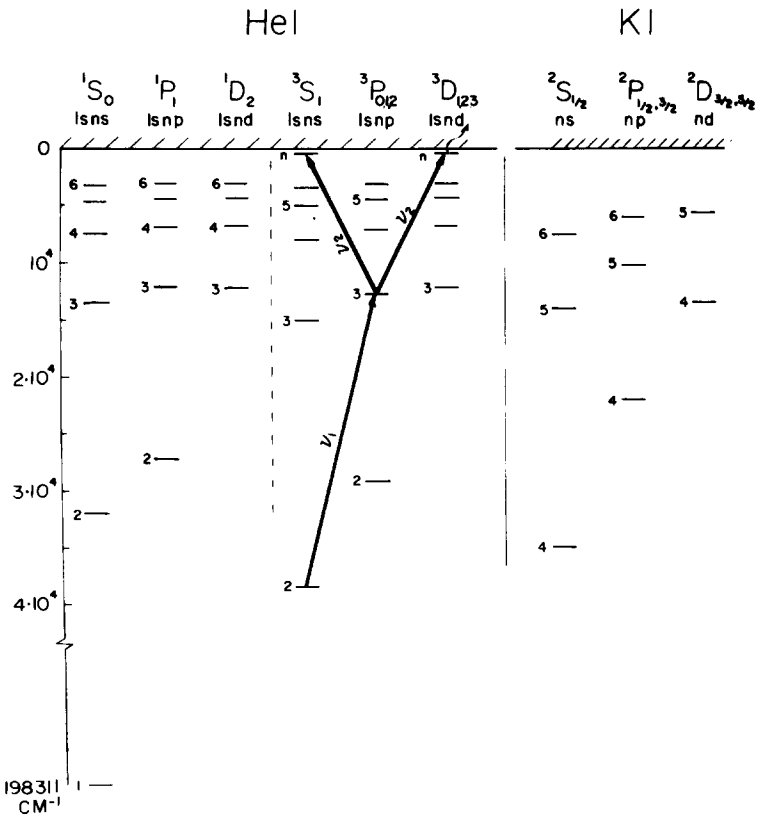

Fig. 1. Energy level diagrams of helium and potassium atoms.

selective laser excitation of the accelerated helium atoms to the $n\,^3S$ and $n\,^3D$ Rydberg states, followed by ionization in an electric field, was carried into effect through the intermediate level $3\,^3P$.

2. Experimental setup

The experimental setup is shown schematically in fig. 2. The continuous helium ion beam was produced by means of a hot-cathode gas-discharge source. The ion energy was 3.9 keV and was determined by the beam-forming electrode potential U_A. The collimated ion beam was then deflected by the deflector d_1 and directed into the charge exchange cell. The deflector is necessary to separate the ion component of the beam from the neutral beam component formed as a result of resonant charge exchange between helium ions and helium atoms in the vicinity of the extracting electrode.

In the charge exchange cell, around 40% of the ions were converted into metastable helium atoms. The fast atoms thus formed passed through the filter capacitor f_1 and entered a field-free region. The filter capacitor field strength was $E_{f1}=9$ kV/cm, and so the helium atoms formed as a result of charge exchange in states with the principal quantum number $n>17$ were ionized and extracted from the beam, as well as the helium ions that failed to exchange their charge. The field-free region with a length of $L=100$ cm was limited by the aperture diaphragms D_3 and D_4. To reduce the residual gas pressure, this region was surrounded by a "jacket" kept at the temperature of liquid nitrogen. The isotope-selective excitation of the helium atoms to the Rydberg states was effected by means of laser radiation at frequencies ν_1 and ν_2 propagating counter to the atomic beam. The ions formed in the field-free region as a result of the fast atoms colliding with the residual gas molecules could be extracted from the beam by the filter capacitor f_2. The highly excited atoms prepared by the laser radiation were ionized in a field ionizer and deflected in the same ionizer onto a detector. The detector could be set at an angle of $\alpha=20°$ or $45°$ to the atomic beam axis. The ionizer was formed by two cylinders 6 mm in diameter spaced at a distance of 14 mm between their axes. With this ionizer system geometry, the electric field along the atomic beam diameter equal to 2 mm was practically constant. Before reaching the detector, the ions passed through the adjustable slit S 0–6 mm in width. The effective neutral atomic beam current was 1–15 nA.

The ions were registered by means of a gated counting system during a time of $\tau=L/v$ equal to the time of flight of the atoms through the field-free region.

The fast atoms were excited with dye lasers pumped by a XeCl excimer laser with a pulse repetition rate of 17 Hz. The lasers operated at 390 and 790–800 nm. The bandwidth $\Delta\nu_L$ of both dye lasers was equal to 0.5 cm^{-1}, and the laser pulse duration was 6 ns.

The natural isotope shift on the first transition $2\,^3S\rightarrow 3\,^3P$ is equal to 1.45 cm^{-1}. As a result of the acceleration of the atoms to an energy of 3.9 keV, it is increased to 7.1 cm^{-1}. In that case, the resonant

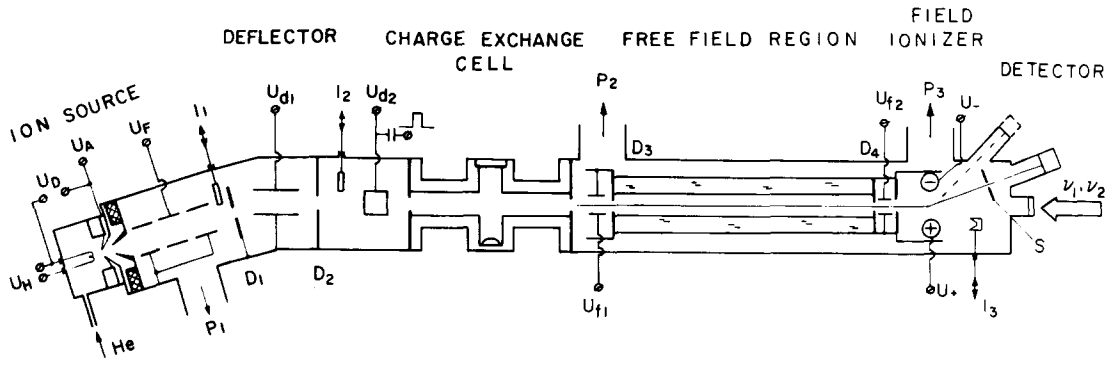

Fig. 2. Schematic diagram of experimental setup: f_1 and f_2 – filter capacitors; $D_1=10$ mm, $D_2=6$ mm, $D_3=6$ mm, and $D_4=2$ mm – aperture diaphragms; d_1 and d_2 – deflectors; I_1, I_2, and I_3 – beam current meters; S – slit.

absorption frequency of the ^{4}He isotope shifts 37.2 cm^{-1} towards the "red" side. The isotope shift of the accelerated atoms at the second step ($3\,^3P \rightarrow n\,^3S$, $n\,^3D$) amounts to around 3 cm^{-1}, which is comparable with the distance between the nS and nD lines. For this reason, to carry out two-step isotope-selective excitation, the frequency of the second-step laser should be chosen so as not to make the stronger nD line of the ^{3}He isotope coincide with the nS line of the ^{1}He isotope. This condition is satisfied, for example, by the $3\,^3P \rightarrow 24\,^3D$ transition.

3. Ionization of fast Rydberg atoms

The fast atoms excited to the $n\,^3S$ and $n\,^3D$ states are ionized in the transverse electric field of the ionizer capacitor. The longitudinal electric field component along the atomic diameter $D_4 = 2$ mm is practically zero. Unlike ions, the Rydberg atoms entering the ionizer begin to undergo the deflecting action of the field only starting from the point they are ionized, hence they are deflected to a smaller angle. With the setting angle of the detector being fixed, to register the signal from the Rydberg atoms, it is necessary to increase the ionizer field strength in excess of that required to deflect ions. Thus, the ionizer capacitor voltage at which the signal due to the ionization of a highly excited state is observed beyond the slit S depends on the distance from the ionizer axis (i.e., the ionizer field strength) at which this state is ionized, and so the transverse electric field ionization system used is dispersive with respect to n.

As with the time-resolved field ionization technique [22], the ionization signal due to the He(nd) states exhibits several peaks corresponding to various projections of the orbital momentum $|m_l|$. More detailed results are presented in ref. [23].

Fig. 3 illustrates the isotope selectivity of the fast atom ionization process. Fig. 3a shows the ion signal as a function of the second-step laser frequency ν_2, the first-step laser frequency being in resonance with the ^{4}He isotope ($\nu_1 = 25671.4$ cm^{-1}). The ion signal for the ^{3}He isotope 10^{-4} in relative concentration (fig. 3b) was obtained with the frequency ν_1 shifted 7.1 cm^{-1} towards the "red" side. As can be seen, the signal from ^{4}He is altogether absent, and the detection selectivity is limited by the background ions at 10^6 at a residual gas pressure of 10^{-7} mm Hg in the system.

The magnitude of the ion background is independent of laser radiation and is directly proportional to

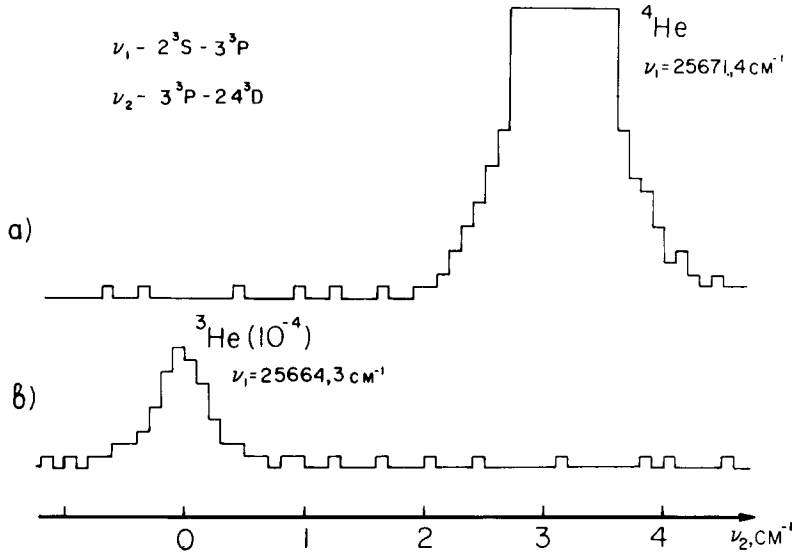

Fig. 3. Ion signal as a function of the second-step laser frequency ν_2: (a) first-step laser frequency $\nu_1 = 25671.7$ cm^{-1} in resonance with ^{4}He; (b) $\nu_1 = 25664.3$ cm^{-1} in resonance with ^{3}He. Averaged over 16 pulses.

the residual gas pressure in the system and the atomic beam current. With the ionization system being adjusted for the level $n=30$ and the residual gas pressure kept at 10^{-7} mm Hg, the background ion counting rate comes to 10^{-3} counts per pulse at an effective atomic beam current of 1 nA. There are two main causes of the formation of background ions. The first is due to the fact that the atoms colliding with the residual gas molecules in the field-free region become excited to Rydberg states:

$$\mathrm{He}(2\,^1\mathrm{S}, 2\,^3\mathrm{S}) + \mathrm{M} \rightarrow \mathrm{He}^*(n) + \mathrm{M}\,.$$

The highly excited atoms thus formed do not differ from those produced selectively by laser radiation and also undergo ionization in the ionizer.

The second cause of the ion background is the ionization of the fast atoms by collision with the residual gas molecules in the region where the Rydberg atoms are subjected to field ionization:

$$\mathrm{He}(2\,^1\mathrm{S}, 2\,^3\mathrm{S}) + \mathrm{M} \rightarrow \mathrm{He}^+ + \mathrm{M} + \mathrm{e}\,.$$

The ions produced in this region are also deflected onto the detector. The effect of these two processes on the magnitude of the ion background can be seen in fig. 4. The arrows indicate the positions of the maxima of signals from atoms excited to states with various n ($l=0$). At a filter capacitor voltage of $U_{f2}=0$ the ion background from the region $n=28$ is due to the above two causes. At $U_{f2}=1000$ V all of the atoms with $n>24$ formed in the field-free region are ionized in the filter capacitor and extracted from the beam, and so the remaining background signal is due to the ionization by collision of the fast atoms in the ionizer region. As seen, the contributions of these two processes are approximately the same, although their cross sections differ by a factor of 500–1000.

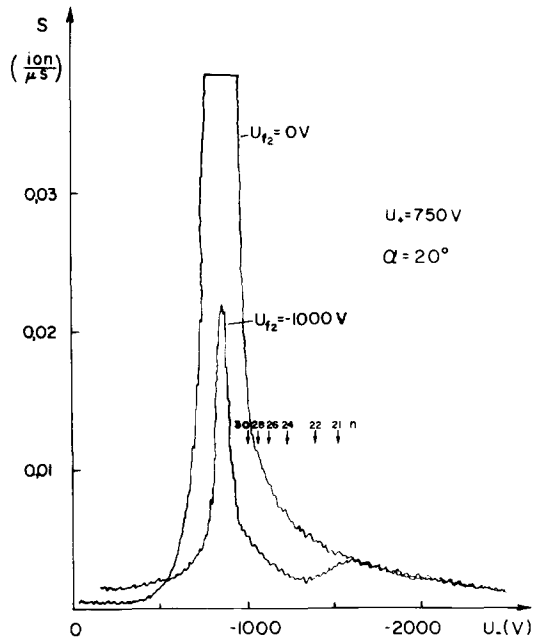

Fig. 4. Background signal as a function of the ionizer capacitor potential U_- at various filter capacitor voltages U_{f2}. The arrows indicate the positions of the maxima of signals from atoms excited to Rydberg states with the principal quantum number n ($l=0$). $P=4\times10^{-7}$ mm Hg.

4. Conclusion

The results of our studies give reason to believe that the multistep collinear laser photoionization of fast atoms is a sensitive and highly selective means for analyzing the isotope composition of substances. Using helium atoms as an example, we demonstrated that the detection selectivity defined as the ratio of the number of the ionized Rydberg atoms of the main isotope, produced by laser radiation, to the number of background ions was 10^6 at a residual gas pressure of 10^{-7} mm Hg in the system. The method makes it possible to detect, without using any additional mass-spectroscopic techniques, ^{3}He with a relative concentration of 10^{-6}. This is of interest in studying the isotopy of natural helium [24].

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